

# Research and Development

CONTROL OF MERCURY EMISSIONS FROM COAL-FIRED ELECTRIC UTILITY BOILERS: INTERIM REPORT INCLUDING ERRATA DATED 3-21-02

# **Prepared for**

Office of Air Quality Planning and Standards

# Prepared by

National Risk Management Research Laboratory Research Triangle Park, NC 27711

# **Foreword**

The U.S. Environmental Protection Agency is charged by Congress with protecting the Nation's land, air, and water resources. Under a mandate of national environmental laws, the Agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. To meet this mandate, EPA's research program is providing data and technical support for solving environmental problems today and building a science knowledge base necessary to manage our ecological resources wisely, understand how pollutants affect our health, and prevent or reduce environmental risks in the future.

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E. Timothy Oppelt, Director National Risk Management Research Laboratory

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# Control of Mercury Emissions from Coal-Fired Electric Utility Boilers: Interim Report

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#### **Abstract**

In December 2000, the U.S. Environmental Protection Agency (USEPA) announced its intent to regulate mercury emissions from coal-fired electric utility steam generating plants. This report, produced by EPA's Office of Research and Development (ORD), National Risk Management Research Laboratory (NRMRL), provides additional information on mercury emissions control, following the release of "Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Plants - Final Report to Congress," in February 1998. The first three chapters describe EPA's December 2000 decision to regulate mercury under the National Emission Standards for Hazardous Air Pollutants (NESHAP) provisions of the Clean Air Act, coal use in electric power generation, and mercury behavior in coal combustion. Chapters 4-9 report: new information on current electric utility fuels, boilers, and emission control technologies; mercury emissions associated with these diverse technology combinations; results and implications of tests to evaluate the performance of mercury control technologies and strategies; retrofit control cost modeling; and mercury behavior in solid residues from coal combustion. The final chapter summarizes current research and identifies future efforts needed to ensure cost-effective control of mercury emissions. References are provided at the conclusion of each chapter.

## **Preface**

This is an interim report, based on data available as of mid-2001, which in some cases are limited. As more data are collected and evaluated, some of the conclusions reached in this report may be modified.

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# Acronyms

ADP Acid Dew Point

AES Atomic Emission Spectrometry
AHC Advanced Hybrid Collector

ASTM American Society for Testing and Materials

CAA Clean Air Act

CCR Coal Combustion Residues

CEM Continuous Emission Monitors

CFBA Circulating Fluidized-bed Adsorber
COHPAC Compact Hybrid Particulate Collector
CS-ESP Cold-side Electrostatic Precipitator

CuCl Cuprous Chloride

CVAAS Cold-vapor Atomic Absorption Spectrometry
CVAFS Cold-vapor Atomic Fluorescence Spectrometry

DI Dry Injection

DOE United States Department of Energy

EPA United States Environmental Protection Agency

EPRI Electric Power Research Institute

ESP Electrostatic Precipitator

ETV Environmental Technology Verification

FBC Fluidized-bed Combustion

FF Fabric Filter

FGD Flue Gas Desulfurization
HAP Hazardous Air Pollutant

Hg Mercury

Hg<sup>0</sup> Elemental Mercury
HgO Mercuric Oxide

Hg<sup>2+</sup> Oxidized or Ionic Mercury
Hg<sub>p</sub> Particle-bound Mercury

Hg<sub>T</sub> Total Mercury

HgCl<sub>2</sub> Mercuric Chloride HgSO<sub>4</sub> Mercuric Sulfate

HS-ESP Hot-side Electrostatic Precipitator

# **Acronyms (continued)**

IGCC Integrated Gasification Combined Cycle

ICR Information Collection Request

kWh Kilowatt Hour LNB Low  $NO_x$  Burner

MC Mechanical Collector

MESA Mercury Speciation Adsorption
MWC Municipal Waste Combustor

MWe Megawatt Electric

MWFA Mixed Waste Focus Area

NESCAUM Northeast States for Coordinated Air Use Management

NETL National Energy Technology Laboratory (DOE)

NO<sub>x</sub> Nitrogen Oxides

OAR EPA's Office of Air and Radiation

OH Method Ontario-Hydro Method

O&M Operation and Maintenance PAC Powdered Activated Carbon

PFF Polishing Fabric Filter

PM Particulate Matter
PRB Powder River Basin
PS Particle Scrubber

PTFE Polytetrafluoroethylene

QA/QC Quality Assurance/Quality Control

 $R_fD$  Reference Dose SC Spray Cooling

SCR Selective Catalytic Reduction

SDA Spray Dryer Adsorber

SEM Scanning Electron Microscope
SNCR Selective Noncatalytic Reduction

# Acronyms (concluded)

TGM Total Gaseous Mercury

UND/EERC University of North Dakota/Energy and Environmental

Research Center

UVDOAS Ultraviolet Differential Optical Absorption Spectroscopy

Wet FGD Flue Gas Desulfurization by Liquid Scrubbing

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# **Executive Summary**

#### Overview

This report documents current knowledge on the emission and control of mercury (Hg) from coal-fired electric utility plants. The purpose of the report is to provide information on the status of government and industry efforts in developing improved technologies for the control of Hg emissions.

This is an interim report, which contains information available in the public domain prior to June 2001. Since then, the results of additional research have been published. This additional information can be found in DOE, EPA, and EPRI reports, in journal articles, and in the proceedings of conferences. Two recent conferences provided significant new information on the control of Hg emissions -- the A&WMA 2001 Annual Conference (Orlando, FL, June 2001), and the A&WMA Specialty Conference on Mercury (Chicago, IL, August 2001).

The first part of the report (Chapters 1 through 3) is directed to readers outside the research community who are interested in Hg emission and Hg control issues. Information is provided on:

- Legislative and regulatory background of EPA's December 2000 decision to regulate Hg emissions from coal-fired electric utility generating stations,
- Studies made in support of EPA's regulatory determination,
- Fuels, combustion technologies, and pollution control technologies used for coal-fired steam electric generating units, and
- Research results from an official Information Collection Request (ICR) on the fuels and technologies used by the utility industry in 1999 at coal-fired steam electric generating stations.

The second part of the report (Chapters 4 through 10) is directed to all readers. It focuses on the review and evaluation of information that has been gathered since the publication of: EPA's Mercury Study Report to Congress; EPA's Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units--Final Report to Congress; and the A&WMA

<u>Critical Review: Mercury Measurement and Its Control.</u> The second part of the report contains information on:

- Hg measurement methods,
- Forms of Hg (speciation) and the capture of Hg in flue gas from combustion of coal,
- Evaluation of the ICR flue gas data on Hg concentrations upstream and downstream of air pollution control devices (APCDs),
- Summary of retrofit control technologies that can be used to limit Hg emissions at coal-fired plants currently equipped with particulate matter (PM) control devices, and dry or wet flue gas desulfurization (FGD) scrubbing systems,
- Estimates of the costs of controlling Hg emissions by the use of powdered activated carbon (PAC),
- Overview of the current coal combustion residue (CCR) management practices and the identification of environmental issues requiring additional research, and
- Conclusions, overview of current research, and research recommendations.

Detailed supporting information is provided in Appendices.

#### **Background**

The 1990 Clean Air Act Amendments required EPA to study the health and environmental impacts of hazardous air pollutants (HAPs) emitted from electric utility boilers. The Agency was also required to conduct a study of the potential health and environmental impacts of Hg emitted from anthropogenic sources in the United States. The EPA subsequently published an 8-volume Mercury Study Report to Congress in December 1997 and a Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units--Final Report to Congress in February 1998. The Hg report to Congress identified coal-fired utility boilers as the largest single anthropogenic source of Hg emissions in the United States. The utility HAP report indicated that there was a plausible link between Hg emissions from coal-fired boilers and health risks posed by indirect exposure to methylmercury.

In December 2000, EPA announced its intent to regulate HAP emissions from coal- and oil-fired electrical generating stations. The decision to regulate HAP emissions from coal-fired units was based on:

- A National Academy of Science study on the health effects of methylmercury,
- The collection and analysis of coal- and flue-gas Hg data under an official Information Collection Request (ICR), and
- Studies concerning the status of Hg emission control technologies.

Three important milestones are incorporated in EPA's decision to regulate HAP emissions from coal-fired electric generating units:

- The proposal of regulations by December 2003,
- The promulgation of regulations by December 2004, and
- Compliance with the regulations by December 2007.

## **Electric Utility Coal Combustion and Air Pollution Control Technologies**

The EPA ICR data collection effort was conducted in three phases. In Phase I, information was collected on the fuels, boiler types, and air pollution control devices (APCDs) used at all coal-fired utility boilers in the United States. In Phase II, coal data were collected and analyzed by the utility industry for 1,140 coal-fired and three integrated gasification, combined cycle (IGCC) electric power generating units. Each coal sample was analyzed for Hg content, chlorine (Cl) content, sulfur content, moisture content, ash content, and calorific value. In Phase III, flue gas Hg measurements were made using the modified Ontario-Hydro (OH) Method for total and speciated Hg. Additional coal samples were collected and analyzed in conjunction with the OH Method measurements.

The EPA ICR data indicated that, in 1999, coal-fired steam electric generating units in the U.S. burned 786 million tons of coal of which about 52 percent was bituminous and 37 percent was subbituminous. Other fuels included lignite, anthracite coal, reclaimed waste coal, mixtures of coal and petroleum coke (pet-coke), and mixtures of coal and tire-derived fuel (TDF). Pulverized coal-fired (PC) boilers represent approximately 86 percent of the total number and 90 percent of total utility boiler capacity. Based on capacity, other types of boilers include cyclone-fired boilers (7.6 percent), fluidized-bed combustors (1.3 percent), and stoker-fired boilers (1.0 percent).

The 1999 EPA ICR responses indicate that a variety of emission control technologies are employed to meet requirements for sulfur dioxide ( $SO_2$ ), nitrogen oxides ( $NO_x$ ), and particulate matter (PM). Most utilities control  $NO_x$  by combustion modification techniques and  $SO_2$  by the use of compliance coal. For post-combustion controls, 77.4 percent of the units have PM control only, 18.6 percent have both PM and  $SO_2$  controls, 2.5 percent have PM and  $NO_x$  controls, and 1.3 percent have three post-combustion control devices.

The different types of post-combustion control devices are listed below:

Particulate matter (PM) control technologies include electrostatic precipitators (ESPs), fabric filters (FFs) (also called "baghouses"), and particulate scrubbers (PS). ESPs and FFs may be classified as either cold-side (CS) devices [installed upstream of the air heater where flue gas temperatures range from 284 to 320 °F (140 to 160 °C)] or hot-side [installed downstream of the air heater and operate at temperatures ranging from 662 to 842 °F (350 to 450 °C)]. Based on current information, it appears that little Hg can be captured in HS-ESPs.

*SO*<sub>2</sub> *post-combustion control technologies* are systems that are classified as wet flue gas desulfurization (FGD) scrubbers, semi-dry scrubbers, or dry injection. Wet FGD scrubber controls remove SO<sub>2</sub> by dissolving it in a solution. A PM control device is always located upstream of a wet scrubber. PM devices that may be used with wet FGD scrubbers include a PS, CS-ESP, HS-ESP, or FF baghouse. Semi-dry scrubbers include spray dryer absorption (SDA). Dry injection involves injecting dry powdered lime or other suitable sorbent directly into the flue gas. A PM control device (ESP or FF) is always installed downstream of a semi-dry scrubber or dry injection point to remove the sorbent from the flue gas.

 $NO_x$  post-combustion control technologies include selective non-catalytic reduction (SNCR) and selective catalytic reduction (SCR) processes. With both of these methods, a reducing agent such as ammonia or urea is injected into the duct to reduce  $NO_x$  to  $N_2$ . SCR operates at lower temperatures than SNCR and is more effective at reducing  $NO_x$ , but it is more expensive.

For PM control, ESPs are used on 84 percent of the existing electric utility coal-fired boiler units, and FF baghouses are used on 14 percent of the utility units. Post-combustion  $SO_2$  controls are less common. Wet flue gas desulfurization (FGD) systems are used on 15.1 percent of the units; and, dry scrubbers, predominantly spray dryer absorbers (SDA), are used on 4.6 percent of units that were surveyed. While the application of post-combustion  $NO_x$  controls is becoming more prevalent, only 3.8 percent of units used either selective non-catalytic reduction (SNCR) or selective catalytic reduction (SCR) systems in 1999.

#### **Mercury Measurement Methods**

When the coal is burned in an electric utility boiler, the resulting high combustion temperatures vaporize the Hg in the coal to form gaseous elemental mercury (Hg $^0$ ). Subsequent cooling of the combustion gases and interaction of the gaseous Hg $^0$  with other combustion products result in a portion of the Hg being converted to gaseous oxidized forms of mercury (Hg $^{2+}$ ) and particle-bound mercury (Hgp). The term *speciation* is used to denote the relative

amounts of these three forms of Hg in the flue gas. The total Hg in flue gas  $(Hg_T)$  is the sum of  $Hg_p$ ,  $Hg^{2+}$ , and  $Hg^0$ . It is the ability to measure these forms of Hg, either collectively or individually, which distinguishes the capabilities of available measurement methodologies.

The Hg in flue gas can be measured by either manual sampling methods or by the use of a continuous emission monitor (CEM). Manual methods are available for the measurement of  $Hg_T$  and the speciation of Hg, including  $Hg_p$ . CEMs are now available to measure gas-phase  $Hg_T$ .

#### Manual Test Methods

Manual sampling methods for measuring  $Hg_T$  from combustion processes are well established. EPA Methods 101A and 29 are routinely used to measure  $Hg_T$  in flue gas from incineration and coal combustion. While a validated reference method for the measurement of the speciated forms of Hg does not exist, the Ontario-Hydro (OH) method is the *de facto* method of choice.

Generally, sampling trains used to collect flue gas samples for Hg analysis consist of the same components: a nozzle and probe operated to extract a representative sample from a duct or stack; a filter to collect PM; and a series of impingers with liquid reagents to capture gas-phase Hg. Sampling trains used for speciation measurements sequentially capture Hg<sup>2+</sup> and Hg<sup>0</sup> in different impingers. After sampling, the filter and sorption media are prepared and analyzed for Hg in a laboratory.

While several research methods exist for performing speciated Hg measurements, the OH Method is presently the method of choice for measuring Hg species in the flue gas from coal-fired utility plants. The OH method has been shown to provide valid Hg speciation measurements when samples are taken downstream of an efficient PM control device. However, the OH Method can give erroneous speciation measurements for locations upstream of PM control devices because of sampling artifacts.

Fly ash captured by the sampling train filter can absorb  $\mathrm{Hg}^{2+}$  and  $\mathrm{Hg}^0$ . Catalytic properties of the fly ash can also oxidize  $\mathrm{Hg}^0$ , resulting in physical and chemical transformations within the sampling train. Transformations caused by the sampling process are called artifacts, and the resulting measurements do not accurately reflect critical properties of  $\mathrm{Hg}$  at the locations where the samples were taken. Sampling methods have not yet been developed to overcome measurement artifacts associated with high flue gas concentrations of fly ash.

#### Continuous Emission Monitors (CEMs)

Continuous emission monitors (CEMs) are in some respects superior to manual measurement methods. CEMs provide a rapid real-time or near real-time response, which can be used to characterize temporal process variations that cannot be measured with manual measurement methodologies. Mercury CEMs are similar to most combustion process CEMs in that a flue gas sample must be extracted from the stack and then transferred to the analyzer for

detection. However, Hg monitoring is complicated by the fact that Hg exists in different forms and that quantitative transport of all forms is difficult.

The CEMs designed to measure total gas-phase Hg ( ${\rm Hg}^{2^+}$  and  ${\rm Hg}^0$ ) are now routinely used in Europe and Japan to measure Hg emissions from incinerators. The Hg concentrations in the stack gas from well-controlled emission sources contain negligible amounts of Hg<sub>p</sub>, and the measurement of gas-phase Hg downstream of the emission control devices can be considered to be equivalent to the measurement of Hg<sub>T</sub>.

The detectors in Hg CEMs typically measure Hg<sup>0</sup> by the use of cold vapor atomic absorption spectroscopy (CVAAS) or cold vapor atomic fluorescence spectroscopy (CVAFS). Hg<sub>T</sub> concentrations are measured by converting (reducing) all of the Hg<sup>2+</sup> in the sample to Hg<sup>0</sup> before it enters the detector. Various conversion techniques exist, including thermal, catalytic, and wet chemical methods. The wet chemical technique is currently used in commercial monitors that are capable of speciation measurement. The use of wet chemical reagents results in high operating costs, which are the primary limitation to the Hg CEM's use as a compliance tool.

Speciating Hg CEMs are highly valuable as research tools. Several commercially available  $Hg_T$  CEMs have been modified to indirectly measure  $Hg^{2^+}$  by determining the difference between gas-phase  $Hg_T$  and  $Hg^0$ . Hg CEMs are susceptible to the same PM-related measurement artifacts associated with manual measurements, and users of Hg CEMs in high dust conditions must consider this problem.

Regardless of the sampling method, the key to reliable and accurate Hg sampling and continuous monitoring is maintaining sample integrity. Flue gases may contain particles that change the species of Hg within the sampling train or CEM system. While this does not change the total Hg measurement, it may bias the determination of Hg vapor species, which may be used to estimate the potential for Hg capture, as well as to assess the performance of control devices. Similarly, common flue gas constituents, such as  $SO_2$ , HCl and  $NO_x$ , may affect quantitative measurement performance.

Additional research is needed to investigate and overcome measurement obstacles so that speciating CEMs can serve as process monitors and as a research tool for evaluating the effectiveness of emission controls. Such research can also provide a better understanding of the factors that affect Hg speciation.

## **Speciation and Capture of Mercury**

## **Mercury Speciation**

The capture of Hg by flue gas cleaning devices is dependent on Hg speciation. Both  $\mathrm{Hg}^0$  and  $\mathrm{Hg}^{2^+}$  are in vapor-phase at flue gas cleaning temperatures.  $\mathrm{Hg}^0$  is insoluble in water and cannot be captured in wet scrubbers. The predominant  $\mathrm{Hg}^{2^+}$ compounds in coal flue gas are weakly to strongly soluble, and the more-soluble species can be generally captured in wet FGD

scrubbers. Both  $Hg^0$  and  $Hg^{2+}$  are adsorbed onto porous solids such as fly ash, powdered activated carbons (PAC), or calcium-based acid gas sorbents for subsequent collection in a PM control device. Hg<sup>2+</sup> is generally easier to capture by adsorption than Hg<sup>0</sup>. Hg<sub>p</sub> is attached to solids that can be readily captured in ESPs and FFs.

Flue gas cleaning technologies that are applied on combustion sources employ three basic methods to capture Hg:

- Capture of Hg<sub>p</sub> in PM control devices;
   Adsorption of Hg<sup>0</sup> and Hg<sup>2+</sup> onto entrained sorbents for subsequent capture in PM control devices: and
- Solvation of Hg<sup>2+</sup> in wet scrubbers.

The factors that affect the speciation and capture of Hg in coal-fired combustion systems include the type and properties of coal, the combustion conditions, the types of flue gas cleaning technologies employed, and the temperatures at which the flue gas cleaning systems operate.

Oxidation reactions that affect the speciation of Hg include homogeneous, gas-phase reactions and heterogeneous gas-solid reactions associated with entrained particles and surface deposits. Suspected flue gas oxidants involved in  $Hg^0$  oxidation include oxygen  $(O_2)$ , ozone  $(O_3)$ , hydrochloric acid (HCl), chlorine (Cl), nitrogen dioxide (NO<sub>2</sub>) and sulfur trioxide (SO<sub>3</sub>). Many of these oxidants are also acid species, which may be significantly impaired by the presence of alkaline species in fly ash, such as sodium, calcium and potassium. Heterogeneous oxidation reactions may be catalyzed by metals such as iron, copper, nickel, vanadium, and cobalt. Conversion of Hg<sup>0</sup> to Hg<sup>2+</sup> may be followed by adsorption to form Hg<sub>p</sub>.

The determination of which mechanisms, oxidants, and catalysts are dominant is crucial in developing and implementing Hg control strategies. For example, the impaired oxidation of Hg in subbituminous coals and lignites is probably related to lower concentrations of HCl in flue gas and high alkalinity of the fly ash. PM collectors and scrubbers reflect this in the low removals of Hg in the ICR database.

#### Fundamentals of Sorption

Sorbents used for the capture of Hg can be classified as Hg sorbents or multipollutant sorbents. Sorbents evaluated for Hg capture have been manufactured from a number of different materials such as lignite, bituminous coal, zeolites, waste biomass, and waste tires. The manufacturing process typically involves some type of thermal treatment. Additives are often used to produce impregnated sorbents.

For coal-fired electric utility boiler applications, the use of sorbents to capture gas-phase Hg (or gas-phase Hg and acid gases) is limited to the use of finely ground powdered sorbents. These sorbents can be injected upstream of PM control devices to collect the sorbent and adsorbed Hg. The development of improved sorbents is needed because of poor sorbent

utilization that results from low flue gas concentrations of Hg and short sorbent exposure times in units equipped with CS-ESPs. The performance of a sorbent is related to its physical and chemical characteristics. The best performing sorbents must be carefully matched to performance requirements as defined by the application for which it is to be used. For example, properties and performance requirements of sorbents used for capture of SO<sub>2</sub> and Hg<sup>0</sup> are quite different. In a similar fashion, the performance criteria for sorbents used with flue gas from bituminous coal will probably be different from the sorbents used with sub-bituminous coals.

Sorbents are porous materials. The most common physical properties related to sorbent performance are surface area, pore size distribution, and particle size distribution. The capacity for Hg capture generally increases with increasing surface area and pore volume. The ability of Hg and other sorbates to penetrate into the interior of a particle is related to pore size distribution. The pores of the sorbent must be large enough to provide free access to internal surface area by Hg<sup>0</sup> and Hg<sup>2+</sup> while avoiding excessive blockage by previously adsorbed reactants. As particle size decreases, access to the internal surface area of the particle increases, along with potential adsorption rates. Powdered activated carbons used for Hg control typically have diameters of 44 µm or smaller.

Mercury can be either physically or chemically adsorbed. Physical adsorption (physisorption) typically results from van der Waals and Coulombic (electrostatic) interactions between the sorbent and the sorbate. The resulting bonds are weak (typically < 10-15 kcal/mole) and are easily reversed.

Chemical adsorption (chemisorption) involves the establishment of a chemical bond (as the result of a chemical reaction, electron transfer). Chemisorption results in stronger bonds than physisorption and is not necessarily reversible. Chemical adsorption is also dependent on the presence of chemically active sites where the sorbate is chemically bound. Some of the chemical constituents of activated carbons influencing Hg capture include: sulfur content, iodine content, and chlorine content. Impregnation of carbons with sulfur, iodine, or chlorine can increase the reactivity and capacity of sorbents. Hg<sup>0</sup> is likely oxidized and sorbed in a rapid two step reaction, either chemically by reaction with strong ionic groups such as  $Cl^-$ ,  $\Gamma$ , or  $S^-$  or physically through interaction with functional groups in sorbent pores.

The  $HgCl_2$  is readily adsorbed onto both carbon and calcium based sorbents, probably by acid-base reactions. Section 5.5 details the fundamental research to develop carbon and calcium sorbents for Hg vapor capture.

#### Evaluation of Sorbents

Sorbents may be evaluated by bench-, pilot-scale, or full-scale experiments. The initial screening of sorbents has typically been conducted using bench-scale, packed-bed experimental reactors. These reactors are used to evaluate the adsorption capacity of sorbents exposed to Hg in a synthetic flue gas made from compressed bottled gases. The reactor is held at a predetermined temperature, and either Hg<sup>0</sup> or HgCl<sub>2</sub> is fed into the synthetic flue gas upstream of

the reactor. An on-line Hg analyzer is used to continuously monitor the Hg content of the inlet flue gas and of that after exposure to the sorbent fixed bed. These reactors are used to determine the effects of temperature and flue gas composition on the performance of sorbents. These reactors provide results that are primarily applicable to the capture of Hg in FF baghouses.

Flow reactors that expose sorbents to flue gas during short residence experiments can be used to simulate conditions associated with ESPs. These reactors can be used to explore the rates of Hg adsorption and determine the effects of temperature and flue gas composition. The most effective screening tests are conducted with reactors that are installed on a slip stream from a pilot- or full-scale coal combustion system. Large pilot- or full-scale tests must be used to assess the effects of mass transfer limitations (i.e., mixing and diffusion of flue gas constituents) and long-term equipment operability.

#### Wet FGD Scrubbers

Oxidized mercury compounds such as  $HgCl_2$  are soluble in water and alkaline scrubbing solutions. Thus, the oxidized fraction of Hg vapors in flue gas is effectively captured when a power plant is operated with wet or semi-dry scrubbers for removing  $SO_2$ . The elemental fraction, on the other hand, is insoluble and is not removed to any significant degree. The challenge to Hg removal in wet FGD scrubbers, then, is to find some way to oxidize the elemental Hg vapor before it reaches the scrubber or to modify the liquid phase of the scrubber to cause oxidation to occur.

## **Evaluation of EPA ICR Mercury Emissions Data**

The methods used to evaluate the ICR data were based on two interrelated objectives. The first method was to estimate the speciated amount and the geographical distribution of national Hg emissions from coal-fired power plants in 1999. The second method was to characterize the effects of coal properties, combustion conditions, and flue gas cleaning methods on the speciation and capture of Hg.

## Mercury Capture by Existing Air Pollution Control Devices

The air pollution control technologies now used on pulverized-coal-fired utility boilers exhibit average levels of Hg control that range from 0 to 98 percent, as shown in Table ES-1. The best levels of control are generally obtained by emission control systems that use FFs. The amount of Hg captured by a given control technology is better for bituminous coal than for either subbituminous coal or lignite.

The lower levels of Hg capture in plants firing subbituminous coal and lignite are attributed to low fly ash carbon content and the higher relative amounts of Hg<sup>0</sup> in the flue gas from combustion of these fuels. The average capture of Hg based on OH Method inlet measurements in PC fired plants equipped with a cold-side ESP is 35 percent for bituminous coal, 3 percent for sub-bituminous coal and near zero for lignite.

Table ES-1. Mean mercury emission reduction for pulverized-coalfired boilers.

Post-combustion Emission Controls Used for PC Boiler		Average Mercury Emission Reduction (%) <sup>a</sup>		
		Bituminous-coal- fired	Subbituminous- coal-fired	Lignite- fired
	CS-ESP	36 %	3 %	-4 %
PM Control	HS-ESP	9 %	6 %	not tested
Only	FF	90 %	72 %	not tested
	PS	not tested	9 %	not tested
PM Control	SDA + ESP	not tested	35 %	not tested
and	SDA + FF	98 %	24 %	0 %
Spray Dryer Adsorber	SDA + FF + SCR	98 %	not tested	not tested
PM Control	PS + FGD	12 %	-8 %	33 %
and	CS-ESP + FGD	75 %	29 %	44 %
Wet FGD	HS-ESP + FGD	49 %	29 %	not tested
System	FF + FGD	98 %	not tested	not tested

a) Mean reduction from test 3-run averages for each PC boiler unit in Phase III EPA ICR data base.

Plants that employ only post-combustion PM controls display average Hg emission reductions ranging from 0 to 89 percent. The highest levels of control were observed for units with FFs. Decreasing levels of control were shown for units with ESPs, PS, and mechanical collectors.

Units equipped with lime spray dryer absorber scrubbers (SDA/ESP or SDA/FF systems) exhibited average Hg captures ranging from 98 percent for units burning bituminous coals to 3 percent for units burning subbituminous coal. The predominance of Hg<sup>0</sup> in stack gas units that are fired with subbituminous coal and lignite results from low levels of Hg<sup>0</sup> oxidization.

The capture of Hg in units equipped with wet FGD scrubbers is dependent on the relative amount of  $\mathrm{Hg}^{2+}$  in the inlet flue gas and on the PM control technology used. Average Hg captures in wet FGD scrubbers ranged from 23 percent for one PC-fired HS-ESP + FGD unit burning subbituminous coal to 97 percent in a PC-fired FF + FGD unit burning bituminous coal. The high Hg capture in the FF + FGD unit is attributed to increased oxidization and capture of Hg in the FF.

Mercury captures in PC-fired units equipped with spray dry scrubbers and wet limestone scrubbers appear to provide similar levels of control on a percentage reduction basis. However, this observation is based on a small number of short-term tests at a limited number of facilities.

Additional testing will be required to characterize the effects of fuel, combustion conditions, and APCD conditions on the speciation and capture of Hg.

#### National Emission Estimates

The data used for estimating the national Hg emissions were: (1) the mean Hg content of coal burned in any given unit during 1999, (2) the amount of coal burned in that unit during 1999, and (3) best match coal-boiler-control device emission factor for the unit. The results of these estimates indicated that:

- Coal and related fuels burned in coal-fired utility boilers in 1999 contained 75 tons of Hg, and
- Forty-eight tons of Hg was emitted to the atmosphere in 1999 from coal-fired utility power plants.

## Multipollutant Controls

The EPA ICR data indicate that technologies currently in place for control of criteria pollutants achieve reductions in Hg emissions that range from 0 to > 90 percent. Current levels of Hg control can be increased by application of retrofit technologies or methods designed to increase capture of more than one pollutant. This multipollutant approach can utilize the synergisms that accrue through the simultaneous application of technologies for  $NO_x$  and Hg control,  $SO_2$  and Hg control, or  $SO_2$ ,  $NO_x$ , and Hg control.

Bench- and pilot-scale tests have shown that Hg capture in PM control devices generally increases as the carbon content of fly ash increases. Increased use of combustion modification techniques that increase ash carbon content will generally increase the amount and capture of  $Hg_p$ .

The EPA ICR data indicate that SCR systems may enhance the oxidation and capture of Hg. Recent pilot- and full-scale tests on bituminous coal-fired units equipped with SNCR + CS-ESP and SCR + SDA/FF systems have confirmed these results. However, improvement in Hg capture appears to be highly dependent on the type of coal burned and the design and operating conditions of SCR systems. The potential in increased Hg capture associated with the  $NO_x$  control system cannot now be quantified. It is believed, however, that the use of combustion modification techniques and post combustion  $NO_x$  control technologies on  $NO_x$  state implementation plan (SIP) units will also increase the capture of Hg in these units.

The retrofit of coal-fired electric utility boiler units to control emission of SO<sub>2</sub> and fine PM is also expected to provide co-benefits in the control of Hg. This is apparent from the increased control of Hg on units equipped with FFs, dry FGD scrubbers, and wet FGD scrubbers. Mercury or multipollutant sorbents will add minimal capital costs to units that are retrofitted with FFs or SDA/FF for control of other pollutants. The use of multipollutant sorbents would be more

costly, but the incremental costs of Hg control would be modest. Technologies designed for use on existing wet FGD units could also be used for new scrubbers that are intended to control SO<sub>2</sub> and the precursors to secondary fine PM.

Generally, the control of Hg emissions via multipollutant control technologies can provide a cost-effective method for collectively controlling the various pollutants of concern.

## **Potential Retrofit Mercury Control Technologies**

A practical approach to controlling Hg emissions at existing utility plants is to minimize capital costs by adapting or retrofitting the existing equipment to capture Hg. Potential retrofit options for control of Hg were investigated for units that currently use any of the following post combustion emission control methods: (1) ESPs or FFs for control of PM, (2) dry FGD scrubbers for control of PM and SO<sub>2</sub>, and (3) wet FGD scrubbers for the control of PM and SO<sub>2</sub>.

## ESP and FF Systems

Least costly retrofit options for the control of Hg emissions from units with ESP or FF are believed to include:

- Injection of a sorbent upstream of the ESP or FF. Cooling of the stack gas or modifications to the ducting may be needed to keep sorbent requirements at acceptable levels.
- Injection of a sorbent between the ESP and a pulsejet FF retrofitted downstream of the ESP. This approach will increase capital costs but reduce sorbent costs.
- Installation of a semi-dry circulating fluidized-bed absorber (CFA) upstream of an
  existing ESP used in conjunction with sorbent injection. The CFA recirculates both fly
  ash and sorbent to create an entrained bed with a large number of reaction sites. This
  leads to higher sorbent utilization and enhanced fly ash capture of Hg and other
  pollutants.

Units equipped with a FF require less sorbent than units equipped with an ESP. ESP systems depend on in-flight adsorption of Hg by entrained fly ash or sorbent particles. FFs obtain the same in-flight Hg adsorption as ESPs and additional adsorption as the flue gas passes through the FF cake.

In general, the successful application of cost-effective sorbent injection technologies for ESP and FF units will depend on: (1) the development of lower cost and/or higher performing sorbents, and (2) appropriate modifications to the operating conditions of equipment being currently used to control emission of PM, NO<sub>x</sub>, and SO<sub>2</sub>.

#### Semi-Dry FGD Systems

SDA systems that use calcium-based sorbents are the most common dry FGD systems used in the electric utility industry. An aqueous slurry containing the sorbent is sprayed into an absorber vessel where the flue gas reacts with the drying slurry droplets. The resulting, particle-laden, dry flue gas then flows to an ESP or a FF where fly ash and SO<sub>2</sub> reaction products are collected.

CFAs are "vertical duct absorbers" that allow simultaneous gas cooling, sorbent injection and recycle, and gas absorption by flash drying of wet lime reagents. It is believed that CFAs can potentially control Hg emissions at costs lower than those associated with use of spray dryers.

Dry FGD systems are already equipped to control emissions of  $SO_2$  and PM. The modification of these units by the use of appropriate sorbents for the capture of Hg and other air toxics is considered to be the easiest retrofit problem to solve.

## Wet FGD Systems

Wet FGD systems are typically installed downstream of an ESP or FF. Wet limestone FGD scrubbers are the most commonly used scrubbers on coal-fired utility boilers. These FGD units are expected to capture more than 90 percent of the Hg<sup>2+</sup> in the flue gas entering the scrubber. Consequently, existing wet FGD scrubbers may lower Hg emissions between 20 and 80 percent, depending on the speciation of Hg in the inlet flue gas.

Improvements in wet scrubber performance in capturing Hg depend primarily on the oxidation of  $Hg^0$  to  $Hg^{2+}$ . This may be accomplished by (1) the injection of appropriate oxidizing agents or (2) the installation of fixed oxidizing catalysts upstream of the scrubber to promote oxidization of  $Hg^0$  to soluble species.

An alternative strategy for controlling Hg emissions from wet FGD scrubbing systems is to inject sorbents upstream of the PM control device. In wet FGD systems equipped with ESPs, performance gains are limited by the in-flight oxidization of Hg<sup>0</sup> and the in-flight capture of Hg<sup>2+</sup> and Hg<sup>0</sup>. In systems equipped with FFs, increased oxidization and capture of Hg can be achieved as the flue gas flows through the FF. Increased oxidization of Hg<sup>0</sup> in the FF will result in increased Hg removal in the downstream scrubber.

# Multipollutant Control Methods

From a long-term perspective, the most cost-effective Hg controls will be those implemented with a multipollutant emission control scheme, wherein Hg sorbents also remove other pollutants, and catalysts and absorbers are employed to remove bulk contaminants such as NO and SO<sub>2</sub>. Mercury is also removed as a consequence of using particular bulk gas sorbents, catalysts, particle collectors, and absorbers. Therefore, while sorbents injected upstream of PM collectors may be readily employed for Hg control, the best long-term schemes will result from

modifying or adding control systems for other pollutants that also control Hg emissions. Chapter 9 discusses several applications under development.

## **Costs of Retrofit Mercury Control Technologies**

Preliminary annualized costs of Hg controls using powdered activated carbon (PAC) injection have been estimated based on recent pilot-scale evaluations with commercially available adsorbents (see Table ES-2). These control costs range from 0.305 to 3.783 mills/kWh, with the highest costs associated with plants having hot-side electrostatic precipitators (HS-ESPs). For plants representing 89 percent of current capacity and using controls other than HS-ESPs, the costs range from 0.305 to 1.915 mills/kWh. Assuming a 40 percent reduction in sorbent costs by use of a composite lime-PAC sorbent for Hg removal, cost projections range from 0.18 to 2.27 mills/kWh with higher costs again being associated with plants using HS-ESPs.

Table ES-2. Estimates of current and projected annualized operating costs for retrofit mercury emission control technologies.

Coal Type (sulfur content)	Existing APCD <sup>a</sup>	Retrofit Mercury Control <sup>b</sup>	Current Cost (mills/kWh)	Projected Cost (mills/kWh)
	CS-ESP+FGD	PAC	0.727 – 1.197	0.436 - 0.718
Bituminous (3% S)	FF+FGD	PAC	0.305 - 0.502	0.183 - 0.301
(070 0)	HS-ESP+FGD	PAC+PFF	1.501 – NA°	0.901 – NA°
	CS-ESP	SC+PAC	1.017 – 1.793	0.610 - 1.076
Bituminous (0.6% S)	FF	SC+PAC	0.427 - 0.753	0.256 - 0.452
(0.070 0)	HESP	SC+ PAC+PFF	1.817 – 3.783	1.090 – 2.270
	CS-ESP	SC+PAC	1.150 – 1.915	0.69 - 1.149
Subbituminous (0.5% S)	FF	SC+PAC	0.423 - 1.120	0.254 - 0.672
(0.070 0)	HESP	SC+PAC+PFF	1.419 – 2.723	0.851 - 1.634

a) CS-ESP = cold-side electrostatic precipitator; HS-ESP = hot-side electrostatic precipitator; FF= fabric filter; FGD = flue gas desulfurization

In comparison, the estimated annual costs of Hg controls, as a function of plant size, lie mostly between the costs for low- $NO_x$  burners (LNBs) and selective catalytic reduction (SCR) systems. The costs of Hg control will dramatically diminish if retrofit hardware and sorbents are employed for control of other pollutants such as  $NO_x$ ,  $SO_2$ , or fine PM.

The performance and cost estimates of PAC injection-based Hg control technologies presented in this document are based on relatively few data points from pilot-scale tests and are considered to be preliminary. However, based on pilot-scale tests and the results of ICR data

b) PAC=powdered activated carbon; SC=spray cooling; PFF=polishing fabric filter

c) NA = not available

evaluations, better sorbents and technologies now being developed will reduce the costs of Hg controls beyond current estimates.

Within the next 2 to 3 years, the evaluation of retrofit technologies at plants where cocontrol is being practiced will lead to a more thorough characterization of the performance and costs of Hg control. Future cost studies will focus on the development of performance and cost information needed to refine cost estimates for sorbent injection based controls, will develop cost estimates for wet scrubbing systems that employ methods for oxidizing Hg<sup>0</sup>, and will determine the costs of various multipollutant control options.

The issue of Hg in residues will also be examined to address concerns related to the release of captured Hg species into the environment. These evaluations will be conducted in conjunction with the development and evaluation of air pollution emission control technologies.

## **Coal Combustion Residues and Mercury Control**

Operation of power plants results in solid discharges including fly ash, bottom ash, boiler slag, and FGD residues. These residues already contain Hg, presumably bound Hg that is relatively insoluble and non-leachable. In 1998, approximately 108 million tons of coal combustion residues (CCRs) were generated. Of this amount, about 77 million tons were landfilled and about 31 million tons were utilized for beneficial uses.

Increased control of Hg emissions from coal-fired power plants may change the amount and composition of CCRs. Such changes may increase the potential for release of Hg to the environment from either landfilling or uses of CCRs. Mercury volatilization from CCRs in landfills and/or surface impoundments is expected to be low due to the low temperatures involved and the existence of relatively small surface area per unit volume of residue. For Hg control retrofits involving dry or wet FGD scrubbers, the residues are typically alkaline and the acid leaching potential of Hg from these residues is expected to be minimal.

There are several commercial uses of CCR where available data on which to characterize the Hg emission potential are lacking. The following CCR uses are given a priority for developing additional data in order to characterize the ultimate fate of Hg:

- The use of fly ash in cement production,
- The volatilization and leaching of residues used for structural fills,
- Leaching of residues exposed to the acidic conditions during mining applications,
- Volatilization of Hg during the production of wallboard from gypsum in wet scrubber residues.
- Mercury volatilization during the production and application of asphalt with fly ash fillers, and
- Leaching or plant uptake of Hg from fly ash, bottom ash, and FGD sludge that are used as soil amendments.

#### **Current and Planned Research**

DOE, EPA, EPRI, the utility industry, and the control technology industry are funding research on the control of Hg emissions from coal-fired boilers. A major portion of this research is being funded under cooperative agreements with DOE. These agreements include cost sharing by EPRI and other industrial partners. Research on these projects is being jointly coordinated under DOE's, EPA's, and EPRI's Hg control technology programs. These research efforts will be used to:

- Develop hazardous air pollution Maximum Achievable Control Technology (MACT) requirements for electric utility generating units,
- Optimize control of Hg emissions from units that must comply with more stringent NO<sub>x</sub> emission requirements under the NO<sub>x</sub> SIP, and
- Develop technologies that can be used to control emissions under multipollutant control legislation options that are currently being considered.

Current research efforts include three full-scale test projects, six pilot-scale test projects on coal-fired units, the evaluation of Hg CEMs, supporting research on the speciation and capture of Hg, and research on CCRs and CCBs. This research includes:

- One full-scale ESP sorbent injection project with tests at four sites,
- One full-scale wet FGD scrubber project at two sites,
- One full-scale project on the effects of SNRC, SCR, and SO<sub>3</sub> conditioning systems at five sites,
- On-going research on the development and use of Hg CEMs,
- On-going speciation, capture, and sorbent development research, and
- Small Business Administration projects on development of sorbents, and measurement methods.

Six new pilot-scale DOE projects have been announced in FY2001. These are:

- Advance particulate collector with sorbent injection (North Dakota-EERC)
- Evaluation of Hg<sup>0</sup> oxidization catalysts (URS Radian Group)
- Spray cooling and multipollutant sorbents (CONSOL)
- Evaluation of multipollutant sorbents and CFBA (SRI)
- Electrical discharge multi-pollution control system (Power Span)
- Evaluation of advanced sorbents (Apogee Scientific)

#### **Conclusions and Recommendations**

Additional efforts are planned to characterize the behavior of Hg in coal combustion systems. Further research is needed on the speciation and capture of Hg and on the stability of Hg in CCRs and residue by-products. Studies on the control capabilities and costs of potential Hg retrofit technologies currently under pilot-scale development are being continued and appropriate control technologies are to be evaluated on full-scale units. Additionally, an evaluation of the co-control of Hg with available PM,  $SO_2$ , and  $NO_x$  controls is needed.

Mercury measurement and monitoring capabilities must be consistent with the regulatory approaches being considered; e.g., speciated vs. total Hg emissions. Field activities need to be coordinated to (1) improve the emissions data base, (2) develop the technologies most appropriate for Agency goals (e.g., Hg-specific vs. multipollutant), and (3) refine cost data and cost-performance models based on actual field experience.

Finally, EPA must continue to work closely with DOE, EPRI and the utility industry to develop Hg and multipollutant control technologies. Collaboration will help ensure that all of the scientific knowledge, engineering skills, and financial resources needed to develop control technologies and establish the most cost-effective regulatory requirements are available.

#### Current and future research should focus on:

- Control of emissions for units with ESPs.
- Control of Hg emissions from subbituminous coals and lignite,
- Evaluation of CFA systems,
- Demonstration of Hg control for units with SDA/ESP and SDA/FF systems,
- Development of Hg<sup>0</sup> oxidizing methods for wet FGD systems,
- Evaluation additives for the oxidization of Hg<sup>0</sup> and the sequestration of Hg<sup>2+</sup> in wet scrubbers,
- Enhancement of fly ash capture by combustion modification techniques,
- Optimization of NO<sub>x</sub> controls for Hg control,
- Control of Hg and other air toxic emissions from units equipped with SCR and wet FGD scrubbers,
- Use and evaluation of Hg CEMs,
- Tests with CEMs to study the variability of Hg emissions,
- Effects of coal blending on Hg capture, and
- Effects of cyclone-, stoker-, and fluidized-bed combustion on Hg control.